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Bibliography on the Application of Reverse Osmosis to Industrial and Municipal Wastewater Research Report No. 18



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**Research Program for the Abatement of Municipal Pollution
under Provisions of the Canada-Ontario Agreement
on Great Lakes Water Quality**

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BIBLIOGRAPHY ON THE APPLICATION OF
REVERSE OSMOSIS TO INDUSTRIAL AND MUNICIPAL WASTEWATERS

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RESEARCH PROGRAM FOR THE ABATEMENT
OF MUNICIPAL POLLUTION WITHIN THE
PROVISIONS OF THE CANADA-ONTARIO
AGREEMENT ON GREAT LAKES WATER QUALITY

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ABSTRACT

This review was undertaken to consolidate some of the available literature pertaining to the various applications of reverse osmosis. The period 1968-1973 (inclusive) was examined in detail.

The references herein emphasize the employment of reverse osmosis in the treatment of municipal and industrial wastewaters. Other areas of application are also included where these may prove useful in defining the limits of this separation technique. Information on related physical separation techniques such as ultrafiltration is also available in the bibliography.

The introductory portion of this report provides sufficient information on the mechanisms and applications of reverse osmosis to permit one to make a general assessment of its suitability. The physico-chemical nature of the reverse osmosis process as well as the membrane types available and their fabrication techniques are examined. An area of importance in the waste treatment field is membrane fouling and appropriate cleaning techniques are summarized for different types of surface inhibition. As different applications are most efficiently handled with appropriate equipment, the various module designs are illustrated.

RÉSUMÉ

La présente étude visait à rassembler quelques-unes des publications techniques disponibles ayant trait aux diverses applications de l'osmose inverse. Elle a porté sur la période de 1968 à 1973 inclusivement.

Les références données mettent l'accent sur l'emploi de l'osmose inverse pour le traitement des eaux usées municipales et industrielles. D'autres domaines d'application sont aussi mentionnés dans les cas où ils pourraient se révéler utiles pour définir les limites de cette technique de séparation. Dans la bibliographie, on trouve aussi de l'information sur des techniques physiques de séparation apparentées, comme l'ultra-filtration.

L'introduction du rapport donne suffisamment de renseignements sur les mécanismes et les applications de l'osmose inverse pour en permettre une évaluation générale des possibilités d'application. On y examine la nature physico-chimique du procédé de l'osmose inverse, ainsi que les types de membranes disponibles et les techniques employées pour leur fabrication. Un aspect important en matière de traitement des eaux usées est le colmatage des membranes; aussi les techniques de nettoyage sont-elles brièvement abordées pour les différents types de colmatage. Étant donné qu'une mise en application efficace nécessite l'emploi du bon matériel, on a ajouté des schémas illustrant différents types de dispositifs servant à l'osmose inverse.

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1. INTRODUCTION

The application of reverse osmosis to the treatment of industrial and municipal wastewaters is one of the many diverse areas in which reverse osmosis is being actively studied and utilized today. The concept of a semipermeable membrane separation process dates back to the exploratory studies of Reid and Breton in the late nineteen-fifties (R6). The first success in developing a suitable membrane came in 1960 from independent research under the direction of S. Sourirajan (S35) who showed that cellulose acetate membranes could be prepared with sufficient solute rejection ability and high flux characteristics to permit their utilization as a viable separation/concentration technique. Subsequent developments in the optimization of membranes from both cellulose acetate and other polymeric materials have enabled routine production of high performance membranes which can be fabricated to function efficiently in a variety of applications. Parallel advances in the design of several large scale membrane modules have permitted reverse osmosis to be effectively employed in many areas.

The most extensive employment of reverse osmosis technology has been in the desalination of brackish water and sea water. Food processing applications have also proven quite successful, particularly in whey separation but also in skim milk utilization as well as the corn, potato and orange processing industries. Concentration of industrial effluent streams for either re-use or final treatment before disposal has proven effective in such widely divergent areas as pulp and paper, organic chemicals, leather tannery wastes, fungicides, pesticides, heavy metals, paints, nuclear wastes, wash waters, and acid mine drainage. The technique has also been successful in the removal and/or concentration of most viruses and bacteria. Finally, reverse osmosis is being carefully evaluated for employment in the treatment of municipal effluent as a tertiary, secondary and even primary renovation technique.

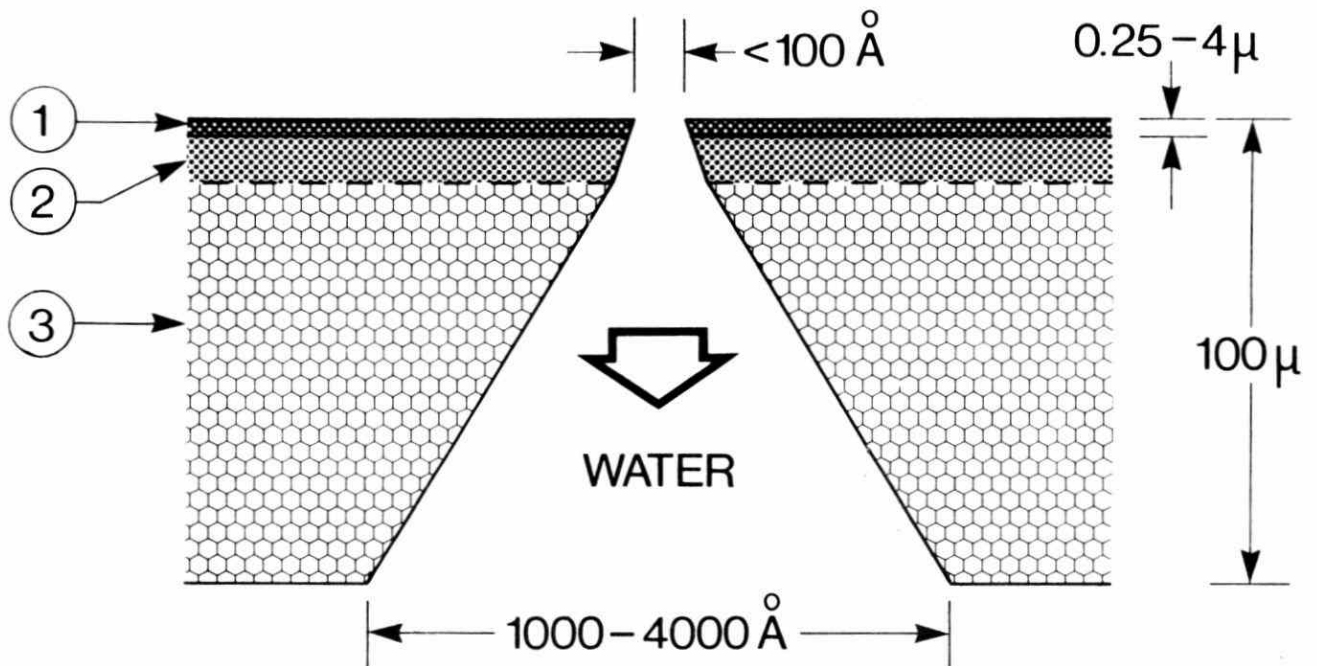
The purpose of this report is to provide a convenient summary up to the end of 1973 of much of the reverse osmosis work being carried out in the waste treatment field. Some references are supplied to other applications of reverse osmosis to permit those unfamiliar with the field to locate basic source material. The preliminary review of the general concepts of reverse osmosis is furnished for the convenience of the reader.

2. REVERSE OSMOSIS

In any system in which two solutions of differing solvent activities are separated by a semipermeable membrane, there will occur a transport of fluid through the membrane in such a manner to produce a thermodynamic equilibrium across the boundary. The pressure head that is built up above, or must be applied to, a solution to prevent the passage of fluid across the membrane is the osmotic pressure, π , and the transfer process is known as osmosis. Osmosis occurs as a consequence of the vapour pressure difference between the solutions and continues until equivalence of the free energies of the two solutions is attained. Reverse osmosis occurs when an external pressure, P , is applied in a sufficient amount to overcome existing osmotic equilibria (i.e. $P > \pi$) and cause the passage of fluid through the membrane in a direction opposite to which transfer would occur in the absence of such an external driving force. On a thermodynamic basis, both osmosis and reverse osmosis involve the transport of a fluid across a membrane in the direction of lowest chemical potential. The application of an external pressure greater than and opposite to the osmotic pressure effectively reverses the direction of fluid transport necessary to satisfy the thermodynamic requirements. Such criteria are unable to specify which component in a solution will be preferentially transported across a membrane, or to explain the mechanism whereby such transport occurs.

3. THEORIES AND MECHANISMS

The operational mechanism of reverse osmosis is generally conceded to be a physico-chemical process in which the nature of membrane-solution interactions largely determines the effectiveness of the technique for the removal/concentration of specific constituents. Ultrafiltration (U.F.), with which reverse osmosis is often compared, is essentially a physical sieving process which separates on the basis of size and shape. While such physical considerations influence the efficiency of reverse osmosis separations, surface interactions are the determining criteria for selective separations of solutes of equivalent size but different chemical natures. Operating pressures are usually significantly different between reverse osmosis and ultrafiltration, the former ranging from about 200-1500 psi and the latter usually being of the order of 100 psi or less. Such pressures are generally indicative of the macroscopic differences between these two membrane types. The sizes of ultrafiltration pores are essentially uniform throughout the membrane, and can have effective diameters that range from 200 Å to 100,000 Å. Reverse osmosis membranes, on the other hand are usually asymmetric (Figure 1), having a macroporous substructure and a dense surface layer. The substructure and the transition region function as effective supports for the dense surface layer. Although there is general agreement that this dense layer is responsible for the separation characteristics of the reverse osmosis membrane, there is still uncertainty about the structure of this surface. Some researchers claim it is uniformly dense and completely amorphous whereas others suggest it contains pores of approximately 10 Å or less. The various proposals concerning surface morphology are almost invariably supported by appropriate theories and transport equations which purport to support the various points of view. Generally speaking, the theories of transport through reverse osmosis membranes may be divided into two main groups, the pore theories and the



- (1) DENSE SURFACE LAYER, $0.25 - 4\mu$
- (2) TRANSITION REGION, INTERMEDIATE DENSITY AND POROSITY; VARIABLE THICKNESS.
- (3) SPONGY POROUS SUBSTRUCTURE

FIG. 1 ASYMMETRIC STRUCTURE OF REVERSE OSMOSIS MEMBRANE.

solution-diffusion theories, although there are many specific differences within each such group.

The originally proposed method of transport across reverse osmosis membranes was due to Reid and Breton (R6) who suggested that two distinct mechanisms occur. Water is considered to be strongly hydrogen-bonded to the carbonyl oxygens in cellulose acetate and is concentrated in the amorphous regions which predominate in such cellulose. Consequently, ions and molecules which can combine with the membrane through hydrogen-bonding and can fit into the bound water structure can be passed across the surface by alignment of hydrogen-bonds in a diffusion controlled mechanism. Non-hydrogen-bonding species are transported by hole-type diffusion through porous regions in the cellulose acetate where there is no tightly bound water. The rates of transport by this secondary sieve mechanism depend mainly on pore size, whereas transport according to the hydrogen-bonding theory depends on the ability to fit into the ice-like membrane structure. Kesting (K10-K12) supports the concept that transport predominantly occurs by a diffusion controlled process governed by the hydrogen-bonding active site theory. Preferential membrane solubility for different compounds exerts a strong influence on such a mechanism and water transport improves when the number of hydrophobic OH sites increases in relation to the number of hydrophobic acetate ester sites. Pores are considered to be membrane imperfections which permit non-selective passage of all components of the appropriate size. Other authors also support the concept of diffusive flow through a pore-free homogeneous phase. Thau, Bloch and Kedem (T3) discounted the pore theory on the basis of the required molecular dimensions for an effective pore, although they qualify their support of the solution-diffusion mechanism with an acknowledgement that no clear distinction may be drawn between diffusion with association and capillary flow. In supporting transport by diffusive flow, Banks and Sharples (B6), suggest increased salt rejection with increase in applied pressure since the pressure effect on the

activity of a component at low concentration is small, and solvent flows increase to a greater extent at higher pressure. Yasuda et al (Y2, Y3, Y5, Y6) present a model based on the free volume concept of diffusive transport which distinguishes between transport by diffusion and bulk flow in homogeneous membranes. All of these transport theories generally characterize the passage of molecules and ions across non-porous membrane boundary layers as occurring through a "solution-diffusion" mechanism. Surface pores are regarded as membrane imperfections and transport through them is considered to be a largely non-selective phenomenon.

The other main group of theories conversely assumes that the surface of reverse osmosis membranes is inherently porous in nature and it is the number and size of these pores as well as the nature of the polymer-solution interactions which control permeability and selectivity. The most acceptable pore theory is that proposed by S. Sourirajan (S33) following his development of the first successful reverse osmosis membrane. He suggests that membrane selectivity is a surface chemistry phenomenon that is dependent on the nature of the specific interactions between the polymer surface and the various components in the solution, as generally outlined in the Gibb's Adsorption Isotherm. Transport through the porous membrane occurs by a capillary flow mechanism, wherein those components preferentially adsorbed at the interface will be transported to the greatest extent. Suitable solutions interacting with appropriate polymers enable the adsorption of a monomolecular layer of a single pure component on the membrane surface. The transport rate is a function of the number and size of the pores on the surface as well as the applied driving pressure. Selectivity is dependent on the thickness of the preferentially adsorbed layer, the size and size distribution of the pores, and the extent of interaction with the surface. This mechanism is generally referred to as the preferentially adsorption-capillary flow mechanism and it assumes all transport occurs through pores and

none occurs through solution in the polymer matrix. The general concepts of this theory have proven compatible with observed permselectivities of both organic and inorganic solutes. Glueckauf (G16, G17) proposed that the preferentially adsorbed layer is less than that suggested by Sourirajan and that the repulsion of ions occurs when solution is enclosed in a narrow pore of material of low dielectric constant. The membrane material is required only to provide suitably sized pores in a matrix of sufficiently low dielectric constant. Salt passage occurs through very large irregular pores. Meares (M37) suggested that although transport occurs predominantly by flow through tortuous channels, some also occurs by homogeneous diffusion in the polymer matrix, rejection of ions in the pores occurring as a result of electrostatic interactions. Although only 1% of the total permeation is proposed to occur by diffusion, approximately half of all solute transport is suggested to occur in this manner. Meares theory is essentially the inverse of that suggested by Kesting and by Reid and Breton. The theories of Glueckauf and Meares offer little explanation of the ability of reverse osmosis to selectively reject non-charged organic species.

Both the foregoing schools of thought, namely the solution-diffusion theory (e.g. Kesting) and the preferential adsorption-capillary flow theory (e.g. Sourirajan) have various strengths and weaknesses. As it is not possible to establish definitively the structure of the surface layer in a reverse osmosis membrane, the application of either approach is dependent on its applicability to the problems at hand and the conceptual simplicity in quantitatively resolving experimental results. Although an exact understanding of the mechanisms of reverse osmosis is not essential for a practical utilization of this process, general understanding of the possible mechanisms may enable wider application of the technique.

4. MEMBRANE PROPERTIES AND FABRICATION TECHNIQUES

Successful application of reverse osmosis requires that efficient membranes be available in suitably designed modules to permit efficient employment in a variety of applications. The optimization of membrane formulation and casting techniques and the search for new and more efficient membrane materials are areas of intensive research. Significant improvements in the performance of existing membrane materials may be achieved by careful control of relevant casting parameters such as humidity, solution temperature, environmental temperature and evaporation time (K44). Small differences in solution composition are also very important. Variation of the solvent composition by less than five percent may result in a reduction in solute rejection of up to thirty times (Johnston, J14) and the type and concentration of the swelling agents employed can cause major differences in membrane performance. Most of these characteristics have been demonstrated in detail for cellulose acetate which is the most explicitly characterized membrane material.

The search for alternate membrane materials has been extensive (Horowitz, H39), but because of the necessity to optimize both the casting solution composition and the casting conditions in order to assess each new polymer adequately, such research is necessarily complex. Several new membrane materials have emerged, the most notable of which is the aromatic polyamide produced by DuPont. A more recent development is an inorganic ultrafiltration membrane (Union Carbide) which may potentially be adapted for reverse osmosis applications. However, the membrane material which produces the highest flux per unit of surface area and has the greatest salt rejection ability is still cellulose acetate.

Although cellulose acetate has excellent permeability characteristics, it is nonetheless limited in certain applications because of its lack of chemical resistivity. The ester groups

tent to undergo hydrolysis in solutions where the pH exceeds 8 or is below 3.5, thereby causing an irreversible reduction in membrane performance. Aromatic polyamide membranes are capable of efficient operation in slightly more basic situations (pH 5 to 11). A material capable of practical utilization over the entire pH range has not yet been developed. Consequently, existing membranes are sometimes employed under conditions in which their practical lifetime is severely limited (Golomb, G28-G30). Even under such non-ideal operating conditions, the process often proves economically advantageous. Both the cellulose acetate and aromatic polyamide membranes must remain moist in order to function efficiently under usual reverse osmosis applications. Recent applications of membrane separation techniques to such areas as helium recovery from natural gas (Habgood, H1) have indicated a need for dry membranes. This has been accomplished with cellulose acetate both by drying existing membranes (Martin, U.S. Patent 3373056) and by modifying casting formulations and techniques to permit the initial formation of dry membranes which may subsequently be repeatedly wetted and dried without adverse effects on their performance (Kesting, K11). Rather than drying the polyamide films, an alternative membrane material has been developed for gas phase separations although at low permeabilities.

Basic differences between the two main membrane-forming polymers have necessitated different fabrication techniques to optimize their reverse osmosis performance. Cellulose acetate films and fibres may be composed of individual polymers or mixtures of polymers in which the acetyl contents may range between approximately 38% and 43%. Molecular weights can vary between about 25,000 and 65,000. Membranes are prepared from a solution consisting of the polymer dissolved in an appropriate solvent (usually acetone) along with a non-solvent (water or formaldehyde) and an appropriate salt, both of which function as swelling agents. Membranes may be cast in sheet or tubular form by physically spreading a desired thickness of this solution

on an appropriate support under controlled atmospheric conditions. Evaporation is allowed to take place for a controlled period of time to permit solvent diffusion through the polymer and from the surface; this results in the characteristic membrane asymmetry. This structure is gelled by immersing the film in a bath of water maintained near freezing. The cellulosic membranes in this "as-cast" condition are generally "macroporous" and show little solute rejection. By heating the membrane in a water bath at an appropriate temperature, the cellulosic material becomes partially crystalline and the resulting polymer shrinkage reduces the size of the pores throughout the film, thereby causing higher solute rejection although at lower permeation rates.

Aromatic polyamide hollow fibres are prepared from a casting dope of the polymer in dimethyl acetamide. The dope is extruded through a sealed, vented insert-spinneret which permits a flow of inert gas into the centre of the filament, thereby maintaining the central bore during fibre setting. Solvent evaporation from the surface (and through the fibre toward the outer surface) occurs at a controlled rate as the filaments pass through a heated cell on emerging from the spinneret. This produces an asymmetric fibre with a dense outer surface and a more porous inner support. The polymer structure is frozen in a quenching bath of water which also serves to leach out extraneous solvent. The fibres are then wound (wet) onto bobbins prior to wrapping the membrane core unit for a completed permeator.

The polyamide used in this process is more resistant to basic solutions than is cellulose acetate, and can operate within the wider pH range of 4 to 11. However, extended usage at more extreme pH's causes irreversible modification of the membrane material and alters its separation characteristics.

5. MODULE CONFIGURATIONS

Once a membrane material has been chosen for a specific reverse osmosis application, the selection of the most appropriate membrane configuration is often critical to the success of the process. Membranes are contained in modules of various designs, the capacity of which can range from a few millilitres per hour to millions of gallons per day. The ability to choose from several module designs permits optimum employment of this separation technique and is partially responsible for the increased utilization of reverse osmosis in recent years. Table I summarizes the membrane and module designs available for cellulose acetate and aromatic polyamides. The addresses of the manufacturers and distributors of appropriate equipment shown in Table I are presented in Appendix I.

There are basically four main types of module designs available: plate and frame, spiral wound, tubular, and hollow fibre. Each design has its specific merits and can consequently achieve optimum efficiency under different conditions of operation. Some general characteristics are described below.

5.1 Plate and Frame

The plate and frame configuration was the first design employed in large scale applications of reverse osmosis membrane technology. Developed by the Aerojet-General Corporation, the units consist of membranes supported on porous plates and arranged one on top of the other (Figure 2). Two different flow configurations may be utilized, one in which the permeated water is collected at the outer edge of the "stack" and the other in which a central collection tube is employed. Turbulence promoting devices adjacent to the membrane-solution interfaces improve transport characteristics and reduce surface fouling to a certain extent.

TABLE I

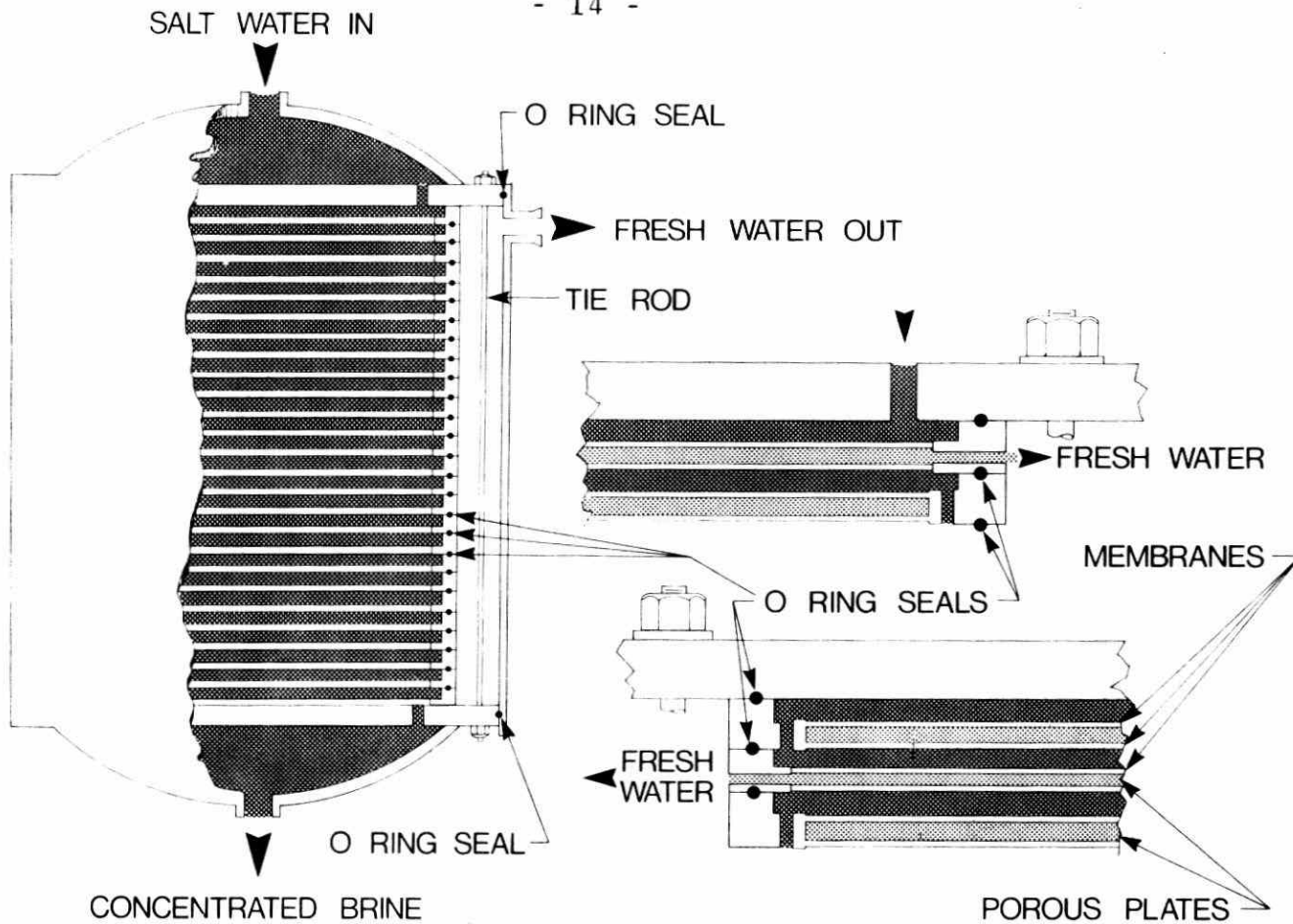
REVERSE OSMOSIS MODULE DESIGNS AND MEMBRANE CONFIGURATIONS

MEMBRANE CONFIGURATION	MODULE DESIGN	POLYMER MATERIAL	MEMBRANE MANUFACTURERS	MODULE MANUFACTURERS & DISTRIBUTERS
Flat	Plate & Frame*	Cellulose Acetate ¹ Aromatic Polyamide ² Inorganic	Envirogenics DuPont Union Carbide	Amicon Gulf Union Carbide
	Spiral Wound	Cellulose Acetate	Gulf General Atomic	Gulf/Ajax/Desalination/ Envirogenics
Tubular	External Tube	Cellulose Acetate Cellulose Acetate	Raypak Universal Water Corporation	Raypak Universal Water Corporation
	Internal Tube	Cellulose Acetate Cellulose Acetate Cellulose Acetate Cellulose Acetate	Universal Oil Products Westinghouse Electrohome Philco Ford American Standard	Universal Oil Products/ Abcor Aqua Chem
Hollow Fibre		Aromatic Polyamide	DuPont	DuPont de Nemours
		Acrylic Cellulose/Cellulose Acetate/Silicone poly- carbonate	Biomed Dow	 Dow/Biorad
		Non-cellulosic	Amicon	Amicon/Mandel Scientific

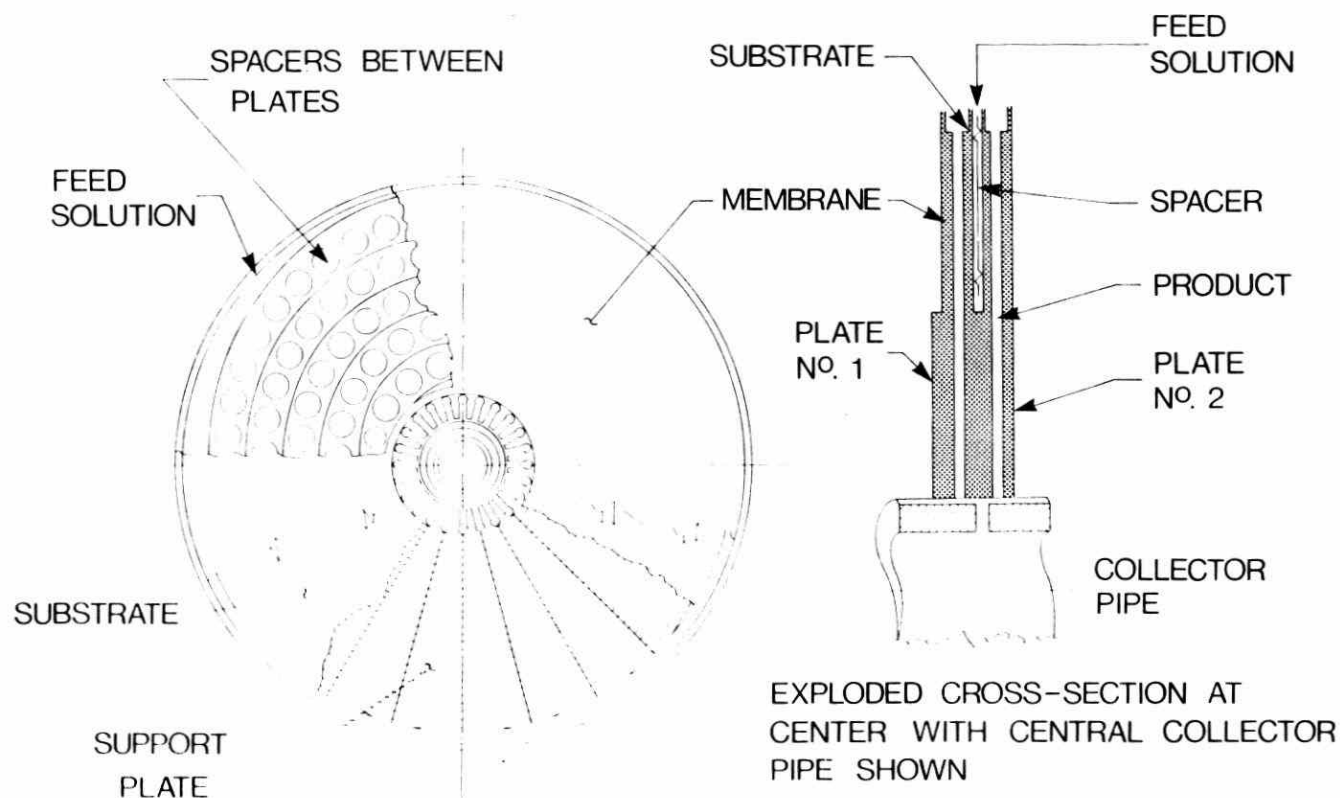
1. Usable pH range 3.5 - 8.0

2. Usable pH range 4.0 - 11.0

* No plate and frame large scale modules are currently manufactured for reverse osmosis units; this type of configuration is a suitable general design for preliminary testing of new membrane materials. Suitable test cells are made by Amicon and used by Dow Chemical for ultrafiltration.



TYPE I DESIGN WITH OUTSIDE COLLECTOR TUBES



TYPE II DESIGN WITH CENTRAL COLLECTOR PIPE

Eastman Chemical Products, Inc.

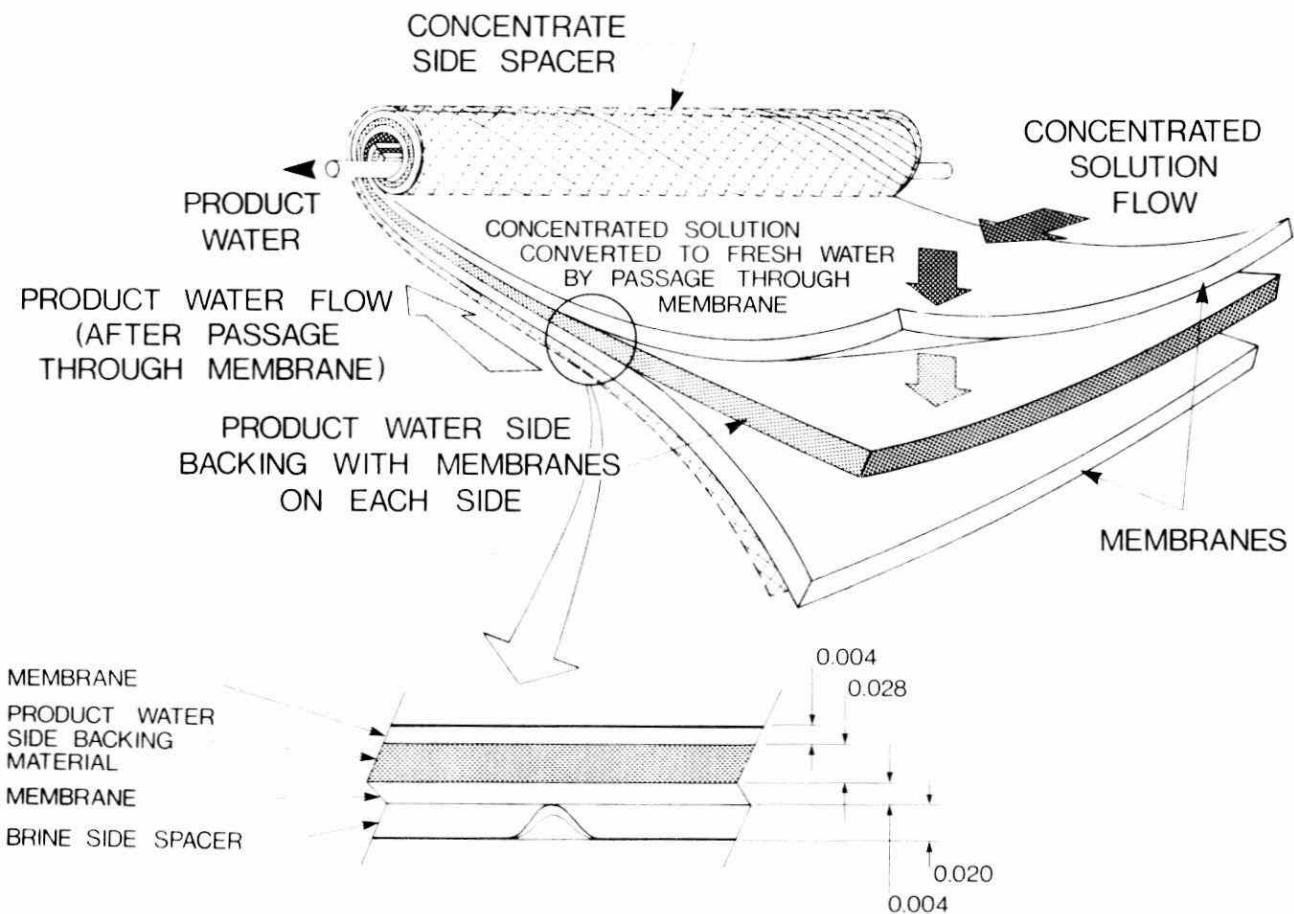
FIG. 2 PLATE AND FRAME REVERSE OSMOSIS UNITS

A limited number of these units were assembled for large scale applications and certain deficiencies became apparent. Because of the requirement for mounting each membrane on an individual supporting device and the necessity of stacking these devices, the required equipment and maintenance costs are excessive in comparison to the productivity obtainable from such an arrangement. The tendency of the membrane surfaces to foul in the narrow separating channels, the large space requirements, and the difficulty of replacing damaged membranes combined to reduce the practicality of the plate and frame configuration. Nonetheless, the units employed in various applications demonstrated that reverse osmosis could be applied to certain large scale processes.

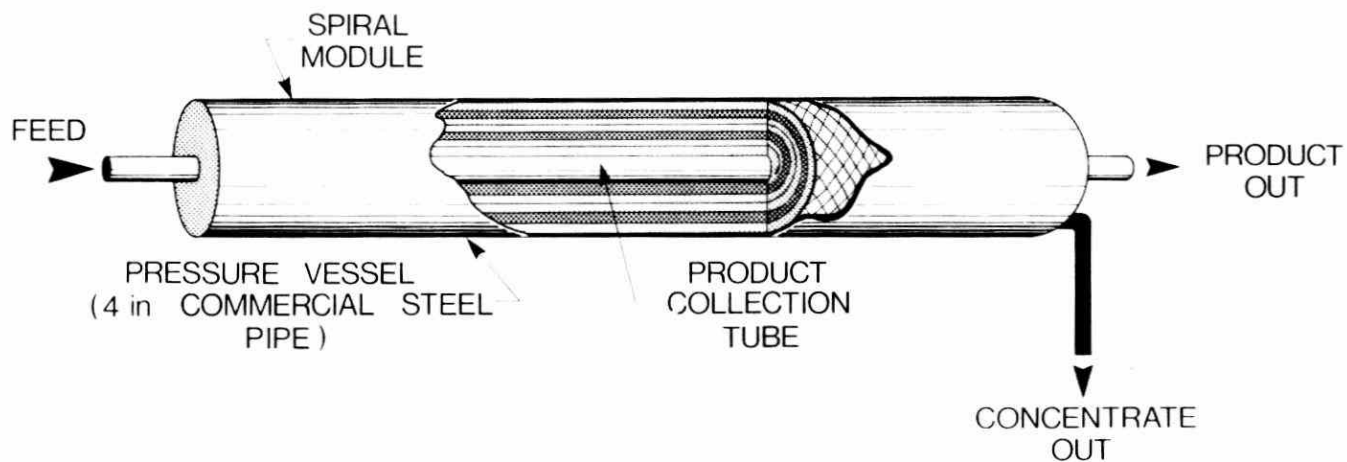
5.2 Spiral Wound

To overcome some of the equipment problems encountered in the plate and frame design, a spiral wound module was developed (Figure 3). The spiral wound configuration consists basically of two large, flat membranes separated by a flexible porous support and wrapped around themselves to form a jelly roll-like structure. The edges of the membrane unit are sealed and the flow of the feed/concentrate solution is directed axially through the roll, thereby encountering the entire surface area before passing out the spacer ports at the end. Permeate passes between the paired membrane layers toward a porous collection pipe at the centre of the wound module. An external tubular pressure container encases the membrane core unit.

This configuration enables maximum membrane film area to be housed in the minimum possible space. Several modules can be arranged in either series or parallel to achieve the desired level of separation and recovery. The restricted nature of the long flow path, however, causes rather substantial pressure drops which may necessitate the use of auxiliary pumps in large scale applications. The proximity of the membrane



SPIRAL-WOUND MEMBRANE MODULE, PARTIALLY UNROLLED.



SCHEMATIC OF SPIRAL-WOUND MEMBRANE MODULE

Gulf General Atomic

FIG. 3 SPIRAL-WOUND REVERSE OSMOSIS MODULE

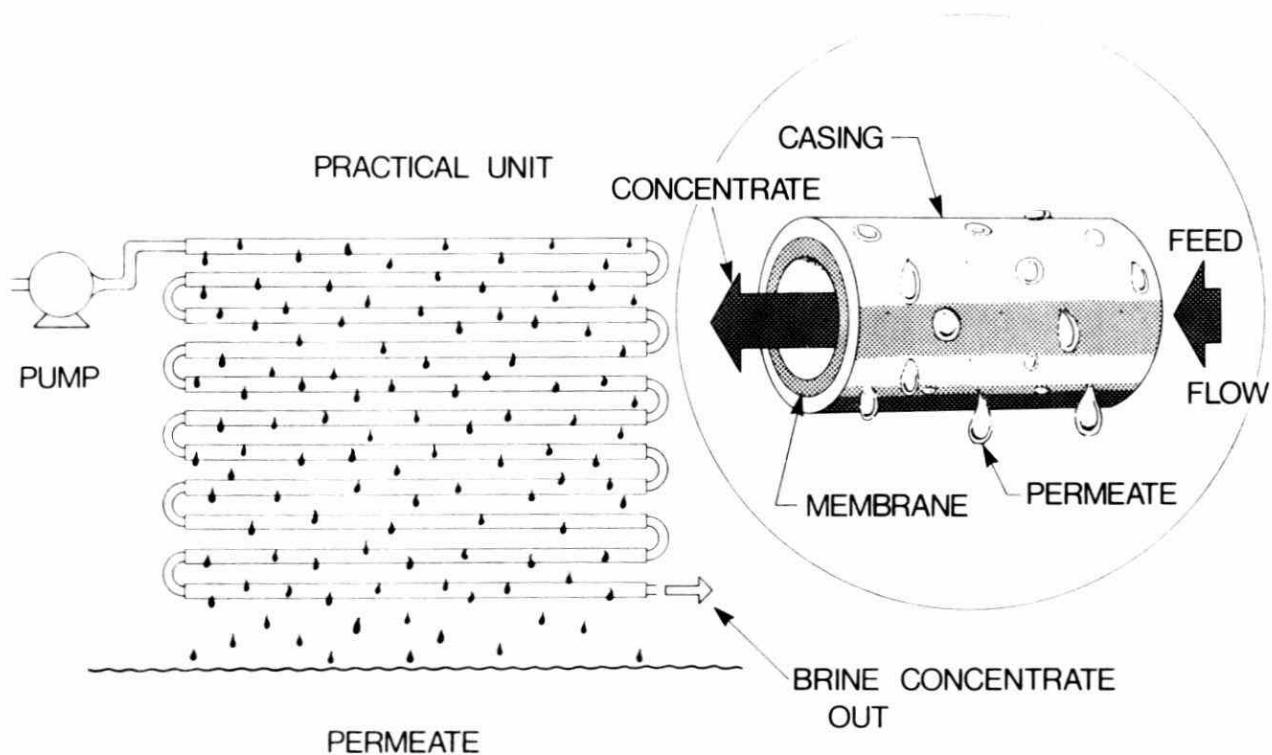
surfaces to each other results in easy membrane fouling and plugging of the narrow interfacial flow channels. Because the surface turbulence is comparable to the plate and frame design, the treatment of viscous fluids or fluids with high solids content is not optimally handled by the spiral wound module.

5.3 Tubular

Modules of the tubular configuration are the most amenable to treatment of viscous liquids. Tubular membranes may be either internally or externally supported (Figure 4) and turbulence promoters can be easily employed with certain designs when required. The ability to adjust flow characteristics through the tubes permits efficient treatment of viscous solutions and reduces surface fouling. Minimum pre-treatment is necessary when using such modules and when required, cleaning by most of the available techniques is readily accomplished. The necessary membrane supports, permeate collection and transfer tubes, module encasing and connecting devices and the small membrane area per module make the tubular arrangement the most expensive to fabricate and install and yield the lowest permeability per unit volume of equipment.

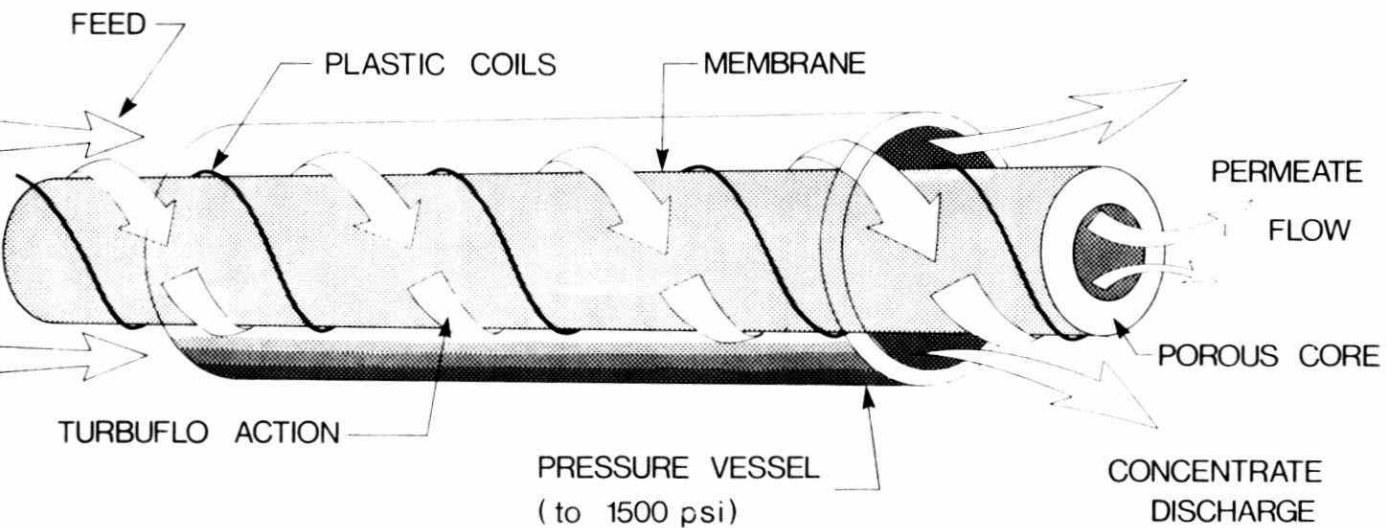
5.4 Hollow Fibre

The hollow fibre design permits maximum membrane area per unit equipment volume. The membranes are spun in the form of hollow fibres, the thickness of which is similar to a human hair (85-200 μ O.D.). These are wrapped around a support frame and the open ends of the looped fibres are potted in a resin matrix. This fibre bundle is encased in a cylindrical container into which the feed solution is pumped under pressure. Permeate is passed through the inside of the hollow fibres to be collected at the open ends encased in the resin (Figure 5).



TYPE I DESIGN WITH MEMBRANE EXTERNALLY SUPPORTED

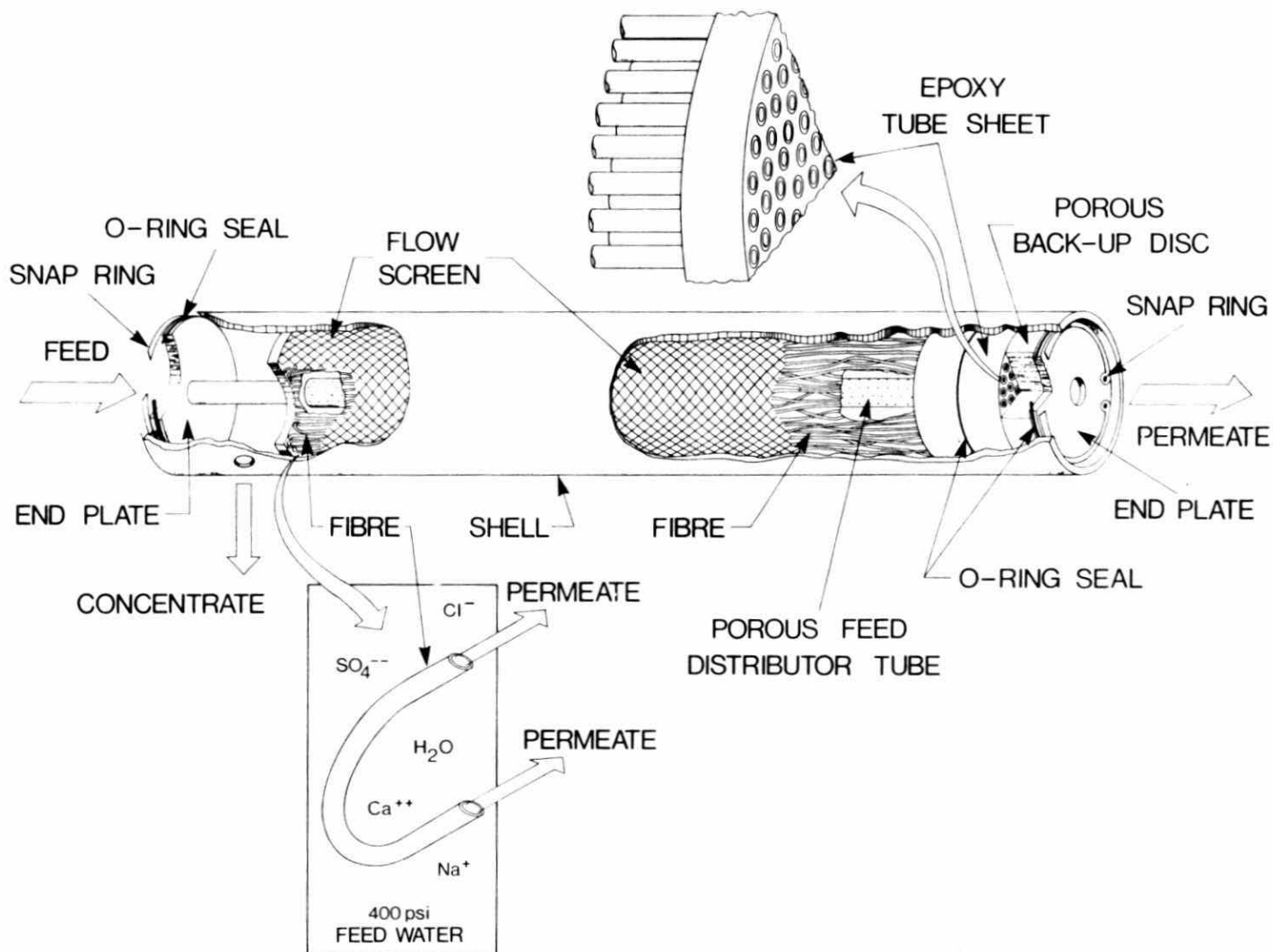
Eastman Chemical Products, Inc.



TYPE II DESIGN WITH MEMBRANE INTERNALLY SUPPORTED

Raypak, Inc.

FIG. 4 TUBULAR REVERSE OSMOSIS



COMMERCIAL PERMEATOR, CUTAWAY DRAWING
Du Pont de Nemours and Co.

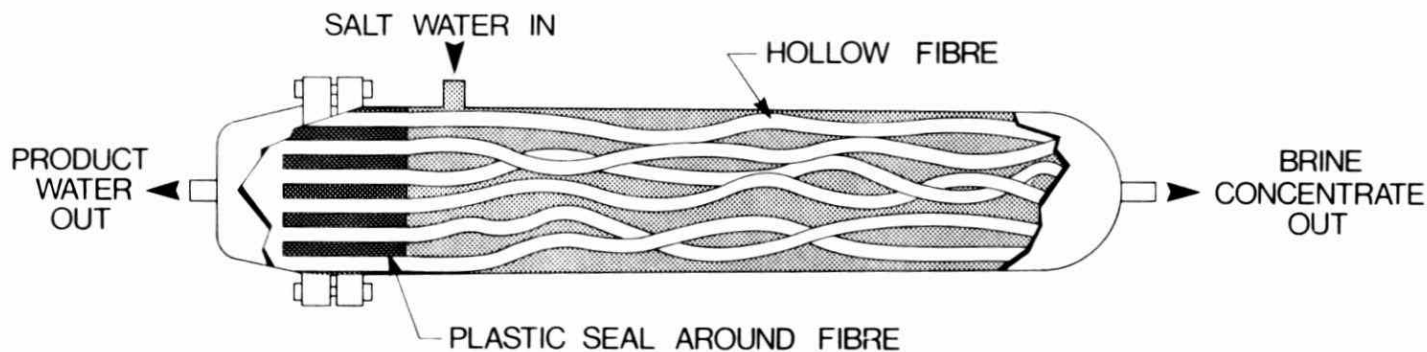


ILLUSTRATION OF HOLLOW FIBRE REVERSE OSMOSIS PROCESS.

FIG. 5 HOLLOW FIBRE REVERSE OSMOSIS MODULE

The main advantage of this configuration is the exceptionally high membrane surface area that is available in very small modules. Although some of the membrane materials are not especially permeable (e.g. aromatic polyamide) on a per unit membrane area basis, the basic membrane density in the fibre-containing modules still results in high capacity units. As the fibres themselves are capable of withstanding the pressures applied, the encasing module is the major mechanical component of the complete unit and no support screens are required. This results in comparatively low equipment costs. Fluids with suspended solids or high viscosities are not usually treated with these modules because of their tendency to foul very easily and the difficulty of adequately cleaning the fibrous surfaces. Pretreatment processes may be employed to render the feed solutions suitable for treatment in such a system. Demineralization of brackish waters and treatment of impure household waters proved to be areas of optimum employment for this module design.

6. MEMBRANE CLEANING TECHNIQUES

In the operation of reverse osmosis units, permeate flux tends to decline as a consequence of fouling of the membrane surface, densification of the polymer substructure due to applied pressures, and irreversible chemical and/or physical damage to the polymer surface layer. In the latter case, hydrolysis of the ester group in a cellulose acetate membrane or physical abrasion of the surface by particulate materials in the feed solution are relevant examples. Surface fouling by colloidal gels or solid precipitates increases the effective osmotic pressure of the feed, thereby reducing the net driving force of the reverse osmosis process. This, in conjunction with reduced permeability due to physical flow restrictions imposed by the adsorbed materials, is one of the major difficulties which must be overcome to successfully apply reverse osmosis to the treatment of various industrial and municipal wastewaters.

In order to justify the use of reverse osmosis in many applications, surface fouling must be easily overcome, either through the development of non-fouling membrane materials or through the optimization of effective cleaning techniques. Membrane fouling depends on feedwater quality, extent of pre-treatment, and the degree of surface turbulence. Five general types of membrane fouling and related problems are summarized below, along with the most appropriate counter measures presently available.

6.1 Membrane Densification and Loose Foulants

(a) Periodical shut down of the unit for several minutes enables the membrane to "relax".

(b) Flush the membrane at twice the normal velocity with feed solution or tap water at reduced pressure.

(c) Air may be introduced into the system to help loosen the foulants during the flushing.

6.2 Hardness Scales

(a) These are precipitates of calcium and magnesium carbonates and sulfates and can be avoided by keeping the water recovery low (<50%), by adding antiscaling solutions such as sodium hexametaphosphate, and by controlling the pH of the solution in the acidic range where precipitation will not occur.

(b) Carbonate scale may resist mechanical flushing but can be dissolved in acid solution. Conversely, calcium sulfate scale is more readily removed by flushing but is less easily dissolved than carbonate.

(c) A warm, weak acid solution (e.g. 1-2% citric acid) or solution of ammonia citrate may be used to remove the scales.

(d) Periodic addition of small amounts of citric acid into the feed help prevent the accumulation of inorganic surface foulants.

(e) The sodium salt of an EDTA solution has also been suggested for CaSO_4 scale removal.

6.3 Iron Oxides and Aluminum Silicates

(a) The treatment and prevention methods are similar to those for hardness scales.

(b) Stronger acids such as HCl in tap water at a pH of about 3 can be circulated through the system for short periods.

6.4 Adsorbed Organics

(a) Fouling by adsorbed organics is sometimes sufficiently weak that circulation of clean water through the system may be adequate to restore membrane performance. However, it is generally necessary to clean the membranes on a regular basis.

(b) Enzyme cleaning is so far the most effective method. An enzyme detergent solution such as Biz, Tergazyme, or Bold at proper concentration and pH is initially circulated vigorously through the system and then the membrane is soaked in the solution for a period of time.

(c) Some organic colloids or proteins could be dissolved by employing pH adjusted water.

6.5 Microbiological Growths

(a) Small amounts of biocide can be introduced into the feed stream or the feed may be presterilized using chlorine or ozone. Free chlorine may damage some membranes, especially the polyamide type.

(b) Methods of cleaning are similar to those employed in treating organic foulings.

(c) Circulation of a solution of sodium hypochlorite through the system for about thirty minutes has also been suggested to sterilize the system.

A physical cleaning method may further be employed in tubular systems. In such a technique, sponge balls with a diameter at least equal to the inside diameter of the tube are pushed through the tube by the applied pressure and are physically able to scrape off precipitants and foulants on the membrane surface as they pass through the system.

7. APPLICATION OF REVERSE OSMOSIS TO WASTEWATER TREATMENT

Reverse osmosis is an effective separation/concentration technique for a wide variety of both organic and inorganic species. It has consequently proved appropriate for the treatment of many industrial and municipal wastes. The more successful employment of reverse osmosis technology has so far been in the area of industrial waste control. This is in part due to the fact that the industrial waste streams are more concentrated and have limited variability in their composition. Appropriate pre-treatment and post-treatment can be more conveniently designed for such systems and disposal/reuse of the concentrate optimized. Successful reuse of waste materials has sufficient merit in some situations to make this treatment process economically advantageous. Reclamation of metals from plating wastes and of whey from cheese processing plants are two examples in which either reuse or the sale and redirection of concentrated components has provided economic impetus to the installation of reverse osmosis units. Other areas in which concentrated wastes can be reused include dilute latex paint and some organic chemical wastes. Various food processing effluents have been successfully treated. Wash water from bean, corn and potato processing operations has been treated, largely to reduce excessively high B.O.D. loadings that result from such industries. Some processing plants can economically employ reverse osmosis as a pre-concentration step in the manufacturing sequence and thereby reduce excessive power costs required for freeze drying (coffee) or evaporation (fruit and vegetable juice) units. Also, petrochemical and leather tannery wastes as well as acid mine drainage can be treated to remove high B.O.D. or toxic materials in order to achieve legislated pollution guidelines. Initial acceptance of reverse osmosis as a method of treating industrial effluents was slow, but many recent practical and economic successes have encouraged rapid development in reverse osmosis for the waste control field (e.g. G30).

Development of higher capacity more efficient modules has prompted several studies to evaluate the efficiency of reverse osmosis as a primary, secondary or tertiary treatment technique for municipal effluent. The ability to remove essentially all virus and bacteria as well as most inorganic and organic species in both dissolved and suspended form suggests this technique may be ideally designed for such effluent treatment. However, high equipment and operational costs, the tendency of the membrane surface to foul, and their susceptibility to shock loadings, as well as the uncertain operational lifetime of the membranes under extreme usage have all been initially cited as major difficulties to be overcome. Reliable data from pilot and full scale test units in this area is rather sparse at present and numerous tests are presently underway. Preliminary results in several areas are encouraging and it appears possible that successful incorporation of reverse osmosis in an advanced municipal waste treatment system may be soon forthcoming.

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SUBJECT INDEX

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SUBJECT INDEX

A. Acid Mine Drainage:

AN4	AN60	H25	K3	K40	L3	M14
M16	N2	S23	W18	W19	W20	

Agricultural Uses: (see also Desalination)

A1	A2
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Anti-Fouling: (see Cleaning)

B. Bacterial: (see Concentration/Separation Processes)

Biomedicine: (see Special Applications)

Boundary Layer: (see Concentration Polarization)

C. Chemical Recovery: (see also Wastewater Treatment)

A7	AN9	B19	D12	G42	G44	K6
P17	P18					

Cleaning: (see also Module Designs/Turbulence Promotors)

B13	B16	B29	B40	C3	C4	F27
G15	H3	J1	M13	M61		

Compaction: (see Membrane Development)

Computer Programs: (see Simulation)

Concentration Polarization:

A10	B35	B40	B44	D13	D17	G20
G40	J8	J9	K16	K38	S12	S13
S17	S46	S56	T5	T6	T7	T13
T14	W14	W25				

Concentration/Separation Processes: (see also Food Processing, Special Applications, Desalination, and Water & Wastewater Treatment)

	C11	D12	K45	L30	S6		
Enzyme, Fermentation	B65	W4					
Protein, Lactose Lactose	B3	B39	B41	B43			
Paint	AN9	AN63	AN65	L14			
Virus, Bacterial	B36 O10 W3	C16 R21	E6 S21	G32 S32	K24 S62	K25 S63	N11 W2

D. Desalination, Demineralization:

A3	A7	AN1	AN3	AN29	AN48	AN62
B4	B33	B42	B45	B47	B54	B55
B61	C1	C26	D2	D3	D18	D21
D22	D23	E1	E10	E12	F5	F17
G15	G19	G33	G34	H30	J10	J11
K5	K17	K30	K44	L6	L10	L15
L16	L27	L34	M2	M8	M15	M16
M36	M47	M60	N14	P7	P8	P11
P13	P14	R1	R11	R12	R17	R18
S4	S7	S11	S16	S53	S54	S56
S59	S65	T9	T11	W7	W8	W14
W26	Y9					

Design: (see Economics and Simulation)

Detergent: (see Sewage Treatment/Nutrients and Phosphates)

Diffusion: (see Theory and Mechanism)

Dye: (see Textile Wastes)

E. Economics: (see also Simulation)

AN11	B7	B23	C11	C12	C20	C37
D12	D25	E11	F3	F4	F5	F7
G30	G38	H11	H12	H36	L2	L15
L16	L17	L22	L33	M15	M56	M70
Q1	R7	R15	W12			

Electron Micrographs: (see Membrane Development)

Electro-osmosis/Electrodialysis:

AN12	AN13	AN23	B45	B58	B60	C14
D11	D22	E1	G35	G37	H15	H36
K30	L3	N12	R3	R22	S20	S62
W9						

Electroplating: (see Metals)

F. Flux Loss: (see Cleaning)

Fouling: (see Cleaning)

Food Processing:

	G41	I1	K14	K19	K29	L45	L47
	M1	M43	M44	M46	P15	P19	P20
	P23	P24	R2	S3	S31	S34	S37
	S61						
Whey	A8 G22 O9	AN24 H33	AN44 L11	AN45 L46	B24 M32	D24 M67	G21 N9
Dairy Products	AN31	B3	B62	N8	N9	P6	
Egg White	L48						
Juice	E5	M45	N13				

Corn	B28			
Maple Sap	AN2	U1	U2	W17
Potato Starch	P26			

G. Gas Concentration/Separation:

A6 H1

Gamma Irradiation: (see Membrane Development)

H. Heavy Metals: (see Metals)

Hollow Fibre: (see Module Designs)

Hyperfiltration: (see also Ultrafiltration)

B4	B50	B62	C7	C25	F25	G7
G8	G38	G39	G40	G45	H20	K9
K35	P7	P8	S12	S13	S14	S17
T5	T6	V3				

I. Industrial Waste Water: (see Wastewater Treatment)

Inorganic Chemicals: (see Metals and Desalination/Demineralization)

Ion Exchange with Reverse Osmosis:

A14 D22 Q1

M. Mathematical Modelling: (see Simulation)

Medical: (see Special Applications and Concentration/Separation)

Membrane Development:

A4	A5	A6	AN10	AN15	AN20	AN22
AN30	AN55	B5	B6	B9	B10	B11
B12	B31	B32	B56	B57	C2	C4
C19	C21	C22	C27	C28	C29	D7
D14	D15	D19	F1	F11	F23	G9
G12	G13	H5	H10	H29	H31	H32
J11	K6	K10	K11	K12	K13	K15
K22	K28	K32	K33	K43	K44	L7
L12	L23	L24	L32	L41	L43	M2
M4	M5	M9	M12	M26	M34	M35
M36	M38	M39	M40	M53	M63	N7
O7	P11	P14	R11	R12	R13	S1
S2	S8	S27	S33	S49	S55	S57
S58	T2	V3	V4	V5	W1	Y1
Y4	Y8					

Membrane Regeneration: (see Cleaning)

Metals:

AN1	AN3	AN4	AN8	AN42	AN68	D22
D23	G19	G28	G29	G30	H13	I6
L18	M42	M62	N1	N10	N12	R19
S29	S36	S38	S39	S40	S41	T1

Microorganism: (see Concentration/Separation and Special Application)

Module Designs :

	AN39	AN47	AN59	B21	B23	D9	F14
	G3	G4	G7	G8	G10	G11	G46
	H14	H19	H20	K1	K2	K17	L36
	L37	L45	M10	M29	M53	M55	R10
	S44	S45	S47	S60	S64		
Hollow Fibre	AN17	AN23	AN57	AN58	AN64	AN69	B8
	B42	C17	C23	C31	C32	C36	C38
	D2	G6	L24	L25	M4	M5	M34
	M35	M41	T1				
Plate & Frame	G46						
Spiral Wound	AN14	AN40	AN54	C29	F19	F21	L8
	L9						
Tubular	C21	F6	F12	F15	G39	H2	R7
	R17	S57	T12	T14	W15		
Turbulence Promotors:	(see also Concentration Polarization)						
	G7	P12	T5	T6	T7	T13	T14

Municipal Waste Treatment: (see Sewage Treatment)

N. Nutrient Removal: (see Sewage Treatment/Nutrient)

O. Organic Removal: (see also Chemical Recovery and Wastewater Treatment)

AN13	AN56	D20	D26	G37	H27	M18
M19	M20	M21	M22	M23	M24	M28
M34	M35	M48	O1	P5	R23	

P. Petrochemical Wastes: (see Wastewater Treatment and Chemical Recovery)

Phosphate Removal: (see Sewage Treatment/Nutrients and Phosphates)

Pilot Plants:

Desalination	B46 F17 R3	B52 F21 S53	B55 H4 S54	D6 M8 S59	D8 M30 W6	E12 M41 W7	F13 R1 W8
Portable/ Mobile	M15	S30	S60	W6			
Shipboard	C1	H35	P13				
Waste Treatment	B60	G43	K4	K23	L9	L26	
Miscellaneous	C12	C35	F17	G23			

R. Radioactive Wastes: (see Chemical Recovery and Wastewater Treatment)

S. Sewage Treatment:

Raw, Primary, Secondary, Tertiary	AN5 B26 C41 G27 K41	AN49 B27 D4 H6 L29	AN50 B31 D16 H7 P3	AN66 B63 E4 I3 S25	B17 C15 F8 J5 T7	B18 C25 G13 J6 W5	B25 C34 G26 K36
Sludge	G35 W5	G47	H7	J5	J6	P3	S25
Nutrients & Phosphates	AN37 K9	B59 O3	C26 R5	C34	H15	H17	K8

Simulation:

AN23	B6	B7	G1	K20	K27	L35
M68	O2	S42	W12			

Special Applications:

B30	B34	B36	B65	C1	C31	C32
G15	G23	G32	G34	G35	G42	H18
H20	H34	H37	H21	K43	L13	L30
M60	P13	R15	R20	S6	S30	S51
S52	V1	W12				

Steel Mill Waste: (see Wastewater Treatment and Metals)

Wastewater Treatment (continued):

D27	D28	E3	E10	F7	F9	F10
F26	G2	G5	G18	G24	G26	G27
G28	G44	H13	H27	H28	I2	I4
K6	K7	K9	K23	K26	K34	K37
K38	K39	K45	L1	L4	L5	L18
L20	L21	L39	L40	L42	L49	M3
M6	M7	M11	M17	M18	M19	M20
M21	M22	M23	M24	M28	M31	M33
M34	M49	M50	M52	M66	M69	N15
N16	O4	O5	O6	P26	R3	R9
R14	R16	S5	S11	S18	S22	S26
S28	S29	S36	S38	S39	S40	S41
S43	S50	S51	T2	T10	T12	W11
W28	Y9					

Water Treatment:

AN41	AN42	AN43	C5	C6	C9	C10
C13	C20	C24	E7	F3	F4	F5
F18	F20	G1	G31	G32	H8	H13
H26	H27	H28	H38	J4	K37	K42
K46	L19	L50	M25	M28	M58	M59
M64	N13	P1	P2	P9	P10	R4
R8	S10	S15	S19	S30	T4	T15
W10	W27	W29				

T. Textile Wastes:

A16 B49 P16

Theory and Mechanism of Reverse Osmosis:

A1	A2	AN25	B5	B6	B20	B33
B38	D7	E9	F23	F24	G9	G16
G17	H9	J3	J13	K5	K33	L1
L4	L5	L43	L44	M20	M21	M22
M23	M24	M27	M37	M51	P4	P5
R6	R13	S4	S35	S55	T3	V2
W1	Y2	Y3	Y5	Y6	Y7	

U. Ultrafiltration: (see also Hyperfiltration)

AN18	AN27	AN63	B1	B2	B3	B4
B34	B36	B38	B39	B43	B48	B62
B64	C16	C33	C38	E8	F2	F16
G13	G21	G36	H7	H18	H29	J2
L14	L30	M31	M54	N11	P6	P17
P18	P20	P21	P22	P23	P24	P25
R14	R21	S6	S9	S21	S24	S31
S32	S42	S63	V1	W3	W4	W5

W. Wastewater Treatment:

A3	A14	AN8	AN9	AN11	AN19	AN26
AN28	AN40	AN49	AN52	AN61	AN64	AN65
B8	B14	B15	B16	B22	B30	B53
C8	C11	C19	C25	C30	C32	C35
C36	C39	C40	D4	D5	D10	D12

APPENDIX I

COMPANIES SELLING COMMERCIAL REVERSE
OSMOSIS OR ULTRAFILTRATION EQUIPMENT

COMPANIES SELLING COMMERCIAL REVERSE OSMOSIS
OR ULTRAFILTRATION EQUIPMENT

1. Abcor Inc., 341 Vassar St., Cambridge, Mass. 02139, USA.
2. Aerojet-General Corporation, Envirogenics Co., 9200
East Flair Drive, El Monte, Calif. 91734, USA.
3. Ajax International, Box 4007, Santa Barbara, Calif. 93103, USA.
4. Ames Crosta Mills (Canada) Ltd., 105 Brisbane Road, Downs-
view, Ontario, Canada.
5. Amicon Corporation, 21 Hartwell Avenue, Lexington, Mass. 02173, USA.
6. Aqua-Chem (Ajax) Inc., 225 W. Grand Ave., Waukesha, Wis. 53186, USA.
7. Aquanomic Inc., 11809 "J" E. Slauson Avenue, Santa Fe Springs,
Calif., USA.
8. Anderson, J.W., Company Limited, Dundas, Ontario, Canada.
9. Better Built Machinery, 441 Market Street, Saddle Brook, N.J.
07662, USA.
10. Benner, R.E., Ltd., 620 Supertest Road, Downsview, Ontario
Canada.
11. Bio-Rad Laboratories, 32nd and Griffin Avenue, Richmond,
Calif. 94804, USA.
12. Burns and Roe Inc., 700 Kinderkamak Rd., Oradell, New
Jersey 07649, USA.
13. Calco, 1120 West Jackson Blvd., Chicago Illinois 60607, USA.
14. Calgon-Havens Systems, 8133 Aero Dr., San Diego, Calif.
92123, USA.
15. Canadian Cabinets Co., 130 Murray St., Montreal 102, P.Q.,
Canada.
16. Carver-Greenfield Corp., 9 Great Meadow Lane, Hanover, N.J.
07936, USA.
17. Chemapec, 1 Newark Street, Hoboken, N.J. 07030, USA.
18. Continental Water Conditioning Corp., Box 26428, 1013
Wall Street, El Paso, Texas 79926, USA.
19. Cosmodyne Corp., 2920 Columbia St., Torrance, Calif 90509, USA.

20. Crane Co., Cochrane Division, 800 3rd Ave., P.O. Box 191,
King of Prussia, P.A. 19406, USA.
21. Crane Canada Ltd., Cochrane Division, 1355 Martin-Grove,
Rexdale, Ontario, Canada.
22. Culligan Canada Limited, Sheridan Park, Mississauga, Ont.,
Canada.
23. Desalination Systems Inc., 1107 West Grant Ave., Escondido,
Calif. 92025, USA.
24. Dorr-Oliver Inc., 77 Havenmeyer Lane, Stamford, Conn.
06904, USA.
25. Dow Chemical Co., Membrane Systems, Barstow Bldg., 2020 Dow
Centre, Midland, Mich. 48640, USA.
26. E.I. DuPont de Nemours & Co., Wilmington, Del. 19898, USA.
27. Eastman Chemical Co., Kingsport Tenn. 37662, USA.
28. Ecotech Inc., P.O. Box 458, Cardiff, Calif. 92007, USA.
29. Electrohome Ltd., 809 Wellington St. N., Kitchener,
Ontario, Canada.
30. Extracorporeal Medical, Royal and Ross Roads, King of Prussia,
Pa. 14406, USA.
31. Foxboro Co., Ltd., 707 Dollard Avenue, La Salle 650, Quebec,
Canada.
32. Gulf General Atomic, P.O. Box 608, San Diego, Calif. 92112, USA.
33. Illinois Water Treatment Co., 840 Cedar Street, Rockford,
Ill. 61105, USA.
34. Ionics, 65 Grove Street, Watertown, Me. 02172, USA.
35. Kaiser Engineers, Kaiser Centre, 300 Lakeside Drive, Oakland,
Calif. 94604, USA.
36. Mandel Scientific Co., Ltd., 4920 De Maisonneuve Blvd.,
Suite 10, Montreal 215, Quebec, Canada.
37. Millipore Ltd., 55 Montpellier Blvd., Montreal 370, Quebec,
Canada.
38. Osmonics, 15404 Industrial Road, Hopkins, Minn. 55426, USA.
39. Osmotic Sciences Div. of Greenwood Industries, 468 Prospect
Street, Newport Beach, Calif. 92660, USA.
40. Peacock Bros., P.O. Box 1040, Montreal Quebec, Canada.

41. Permutit Co., E49 Midland Avenue, Paramus, N.J. 07652, USA.
42. Pharmacia Co., 110 Place Cremazie, Suite 412, Montreal 351,
Quebec, Canada.
43. Philco Ford, Ford Road, Newport Beach, Calif. 92633, USA.
44. Polymetrics, 3011 Corvin Drive, Santa Clara, Calif. 94051, USA.
45. Raypak Inc., 31111 Agoura Road, Westlake Village, Calif. 91361, USA.
46. Raypak Thermonics (Canada) Ltd., 120 Skyway Avenue, Rexdale,
Ontario, Canada.
47. Schleicher & Schuell, Keene, N.H. 03431, USA.
48. Selas Corp., Spring House, Penn. 19477, USA.
49. Triton Water Systems, Inc., 15600-01 So. Main Street, Gardena,
Calif. 90248, USA.
50. Ultradynamics Corporation, 2 Wait Street, Paterson, N.J.
07524, USA.
51. Universal Water Corp., 11722 Sorrento Valley Road, San Diego,
Calif., USA.
52. Ultrascience, 2504 Gross Point Road, Evanston, Ill. 60201, USA.
53. Vaponics Inc., Cordage Park, Plymouth Mass. 02360, USA.
54. Westinghouse Canada Limited, Box 510, Hamilton, Ontario, Canada.
55. Westinghouse Electric Corporation, Power Generation Divisions,
Heat Transfer Division, Lester Branch, P.O. Box 9175,
Philadelphia, Penn 19113, USA.